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Original article

Effect of loading conditions on the fracture toughness of zirconia

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Abstract

Purpose: A Vickers hardness indenter was pressed into yttria-stabilized zirconia (Y-TZP) by the indentation fracture method (IF method).
Methods: The effect on the calculated Vickers hardness, fracture toughness values, and indentation fracture load (9.8, 49, 98, 196, and 294 N) was examined to deduce the optimum conditions of the IF method. Calculated Vickers hardness and fracture toughness values were analyzed with one-way analysis of variance and then multiple comparisons (Scheffe). The appearance of on indentation and cracks was also evaluated using a scanning electron microscopy (SEM).
Results: Indentation of Y-TZP was generated by 9.8 and 49 N of indentation fracture load, however cracks could not be confirmed with the microscope attached to the Vickers hardness tester. Both indentation and cracks were observed at 98, 196 and 294 N of indentation fracture load obtained values of 7.1 and 6.8 MPam$^{1/2}$. Cracks noted at the 98 N were not clear, whereas the 196 and 294 N showed especially clear cracks. Due to the hardness of zirconia and the light loads, fracture toughness values for 9.8, 49, and 98 N could not be calculated. There was no significant difference between 196 and 294 N, when calculated fracture toughness values were analyzed with multiple comparisons. SEM revealed clear indentation and cracks, that extended linearly, but no chips or fractures were observed. Surface changes were observed at 196 and 294 N that are presumed to be accompanied by phase transition around the cracks.
Conclusions: Optimum experimental conditions of the indentation fracture load in the IF method were determined as 196 and 294 N.

Keywords: Zirconia; Fracture toughness; Indentation fracture method; Vickers indenter; Vickers hardness

1. Introduction

In recent dental therapy, demands for esthetics and biocompatibility of restoration devices using biomaterials are increasing [1–5]. Zirconia that is increasingly applied to a clinical setting is a biomaterial with excellent strength and toughness [6–10]. Zirconia gathers attention as a material that has functions and strength as well as, or better than, metal frames because zirconia is available for use as a substructure or framework material for the application of porcelain in the fabrication of all ceramic crowns and bridges [11–17].

When stress is loaded, ceramics generates very small deformation and results in fracture before plastic deformation. Considering that strength of solid substance is a required stress per unit area up to the point of fracture, area is proportional to required stress up to the point of fracture in metals which are ductile materials; however, such a similarity rule does not come into existence in brittle materials. Therefore, breaking strength evaluation covering brittleness of ceramics is required [18–25], and ISO [26] and JIS [27] standards describe fracture toughness value testing methods for advanced ceramics. However, it has been known that single-edge precracked beam (SEPB) method that is used for these standards is difficult to give experimental cracks, and that it is required to obtain Young's modulus from different experiments. Moreover, different correction factors are used for fracture toughness values obtained for commercially available porcelains by different researchers, and this difference affects test conditions including test load, loading time, and loading rate. Thus, the indentation fracture (IF) method has always been applied as a fracture toughness value testing method in the field of dentistry because test slip

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preparation and the test method are easy and simple. IF method is a method to calculate fracture toughness value based on the lengths of impressions and cracks generated by indentation of a Vickers indenter into mirror polished test specimens. On the other hand, the IF method has characteristics that fracture toughness values cannot be calculated accurately when the load is too heavy because interfacial fractures occur on the test specimens, as well as when the load is too light because insufficient cracks occur [19,23,24]. Therefore, an optimum indentation fracture load should be selected for each different material to be measured.

Zirconia produces about 4% of volume expansion by crystal phase transformation that is caused by transformation of tetragonal crystal stabilized at room temperature to monoclinic crystal by loading. Therefore, development of cracks is prevented because crystal phase transformation from tetragonal crystal to monoclinic crystal accompanied by volume expansion as well as compression stress at the leading end of the cracks are occurred when cracks are grown in zirconia. It is required to evaluate material characteristics of the new material, including how loading changes indentation and crack, and how much the fracture toughness value is improved as a result of the loading in zirconia, which is a high-strength ceramic. It would contribute to clinical application, which has a demand for safety and assurance, if measure of fracture toughness values for such high-strength zirconia would be determined.

In this study, effects of the indentation fracture load on indentation formed by Vickers indenter, crack length, and fracture toughness values were examined using the IF method as a means to evaluate brittleness with the aim of normalization of measuring conditions for zirconia.

2. Materials and methods

2.1. Materials

Zirconia, 3 mol yttrium-stabilized tetragonal zirconia polycrystal (3 mol Y-TZP) (Kavo Everest® Zirconium Soft, Kavo, Biberach, Germany), of composition 5.0 wt% Y_2O_3 to 94.8 wt% ZrO_2, was used as a material of the experiment. 3 mol Y-TZP is usually using the present clinical scene.

2.2. Specimen preparation

Test specimen was prepared at a size of 2 mm (width) × 5 mm (thickness) × 25 mm (length) using a block of semi-sintered body of 20 mm (width) × 20 mm (thickness) × 40 mm (length) (Kavo Everest® Zirconium Soft, Kavo), which was dimensionally corrected with 20.19% of shrinking percentage, by cutting with a low-speed cutter (ISOMET, BUEHLER), followed by calcining with a baking furnace (Kavo Everest® therm) at 1450 °C as a final heating temperature for 10 h according to conditions specified by the manufacturer. End faces of the calcined test specimen were processed in parallel (0.05 mm). Furthermore, the surface of the test specimen was polished with #150, #400, #600, #1200, and #2000 grit waterproof abrasive papers, and then was mirrored with a dedicated buff using 1.0 μm diamond paste.

2.3. Test conditions

Experiment was conducted with the IF method using a Vickers hardness tester (AVK-A, Akashi, Kanagawa, Japan) by pressing a Vickers indenter into the test specimen to generate semicircular or semielliptical, vertical crack around the indentation. Length of this crack was measured, and fracture toughness value ($MPam^{1/2}$) was calculated for the measured value obtained using Niihara's formula [23] as below:

$$K_{IC} = 0.203(c/a)^{-3/2}Ha^{1/2}$$

K_{IC}, fracture toughness value ($MPam^{1/2}$); a, 1/2 of indentation diagonal length (μm); c, 1/2 of crack length (μm); and H, Vickers hardness (Hv).

Meanwhile, five conditions of indentation fracture load, 9.8, 49, 98, 196 and 294 N (1, 5, 10, 20 and 30 kgf), were used to examine the effect of difference of loading on fracture toughness value; and load holding time was set to 15 s. Experiments were conducted 10 times for each condition, i.e., total 50 times. Calculated Vickers hardness and fracture toughness values were analyzed with one-way analysis of variance and then multiple comparisons (Scheffe). The significance level was defined as 95%.

2.4. Scanning electron microscope observation

Test specimens after a test using the IF method were observed using a scanning electron microscope (SEM, S-4000, Hitachi, Tokyo, Japan) to examine the appearance of indentation and cracks.

3. Results

3.1. Indentation diagonal lengths, crack lengths, Vickers hardness, and fracture toughness value

Fig. 1 shows indentation diagonal lengths and the effect of indentation fracture load. Average of indentation diagonal lengths (a) occurred by 9.8, 49, 98, 196 and 294 N of indentation fracture load were 15 ± 1.3, 39 ± 0.4, 56 ± 0.4, 81 ± 0.4, and 101 ± 0.7 μm, respectively. The indentation diagonal length was increased with increase of the loading. Correlation between the loading and the indentation resulted in a correlation coefficient of $r = 0.98$ and a regression formula of $y = 2.81x + 21.57$; and was highly significant.

Fig. 2 shows the effect of the indentation fracture load on crack length. Crack lengths (c) at 9.8 and 49 N were not measurable because the cracks were too small to measure with the measuring microscope of the Vickers hardness tester. Average crack lengths (c) at 98, 196 and 294 N were 112 ± 4.4, 192 ± 8.5, and 260 ± 4.6 μm, respectively. The crack length was increased with increase of the loading. Correlation between the loading and the crack length resulted in a correlation

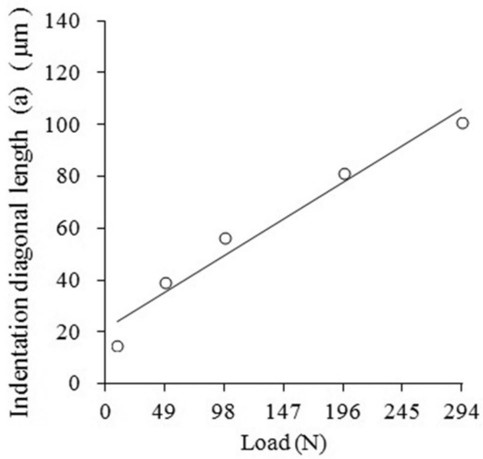

Fig. 1. Effect of indentation load on indentation diagonal length.

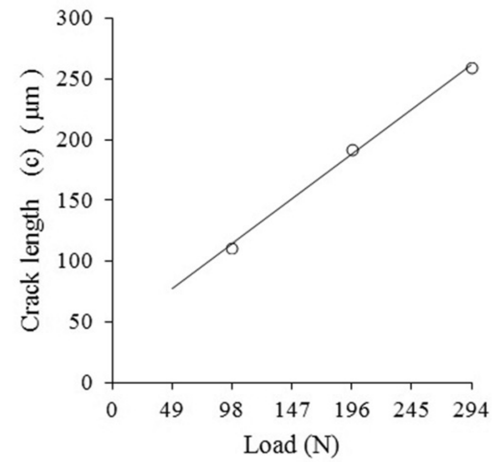

Fig. 2. Effect of indentation load on crack length.

coefficient of $r = 0.99$ and a regression formula of $y = 7.39x + 40.53$; and was highly significant.

Fig. 3 shows the effect of the indentation fracture load on Vickers hardness. Average Vickers hardness (Hv) occurred by 9.8, 49, 98, 196 and 294 N of indentation fracture load were 2073 ± 306, 1518 ± 28, 1459 ± 18, 1400 ± 14, and 1371 ± 17 Hv, respectively. The Vickers hardness was decreased with increase of the loading. Correlation between the loading and the Vickers hardness resulted in a correlation coefficient of $r = -0.65$ and a regression formula of $y = -17.76x + 1798.53$; and was significant.

Fig. 4 shows the effect of the indentation fracture load on fracture toughness value. Average fracture toughness value (K_{IC}) occurred at 9.8 and 49 N cannot be calculated because the crack lengths were not measurable. Average fracture toughness values at 98, 196 and 294 N were 8.0 ± 0.5, 7.1 ± 0.5, and 6.8 ± 0.2 MPam$^{1/2}$, respectively. The fracture toughness value was decreased with increase of the loading. Correlation between the loading and the Vickers hardness resulted in a correlation coefficient of $r = 0.68$ and a regression formula of $y = -0.06x + 8.50$; and was significant. In Scheffe's multiple comparison, there were significant differences between 98 and 196 N as well as between 98 and 294 N with 1% level of significance, and were highly significant. However, there was no significant difference between 196 and 294 N.

3.2. Scanning electron microscope observation

Fig. 5 shows SEM images of indentation and crack. At 9.8 N, indentation was very small, and the crack was immeasurable. At 49 N, a crack that cannot be confirmed with the measuring microscope of the Vickers hardness tester was observed. At 98, 196 and 294 N, both indentation and cracks can be clearly observed.

In addition, at 196 and 294 N, surface changes that are presumed to be accompanied by phase transition were observed in process zone wake around the crack. In all conditions producing cracks, the cracks presented fine linear appearance, and no surrounding fracture due to chipping was observed.

4. Discussion

4.1. Effect of indentation fracture load on fracture toughness value of zirconia (optimum condition of the IF method)

When a Vickers indenter is pressed onto the surface of a brittle solid, a semicircular or semielliptical, vertical crack is generated around the indentation. The IF method is a test to evaluate breaking strength of ceramics using a Vickers hardness tester based on the size of this crack [26–30]. Indentation fracture load and load holding time are adjustable to 9.8, 49, 98, 196, 294, and 490 N and to 1–30 s, respectively, in the Vickers hardness tester used in this study. At first, it is required to set the load holding time at a certain length to examine the effect of the indentation fracture load. According to a report by Okada et al. [24], in which fracture toughness value in porcelain was evaluated, 5 s of load holding time was not enough to form stress to develop a crack adequately, and that there was no difference between 15 s and 30 s. Accordingly, we selected 15 s as a load holding time to improve the efficiency of our experiment. A crack generated by indentation fracture load of 490 N is produced beyond the range of the viewing field of the measuring microscope of the Vickers hardness tester; therefore, we excluded the condition preliminarily.

As a result of the experiment, indentation was generated by 9.8 and 49 N of indentation fracture load; however, the cracks cannot be confirmed with the microscope attached to the Vickers hardness tester. Both indentation and cracks were observed at 98, 196 and 294 N of indentation fracture load, and those are especially clear at 196 and 294 N. Also, according to Japanese Industrial Standards for Vickers hardness (JISZ2244), it is recommended that measurement should be conducted within the range from 25 to below 75% of the viewing field of the measuring microscope of the Vickers hardness tester. The diameter of the viewing field of the attached microscope used in this study was 916 μm. Thus, 25–75% of the viewing field of the microscope is 229–687 μm. According to our experimental

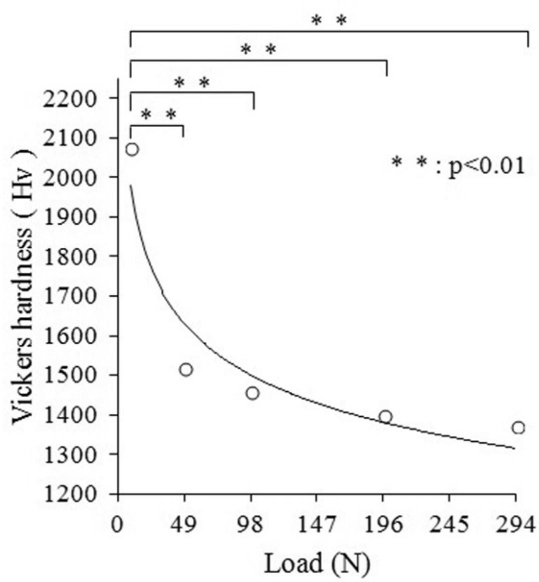

Fig. 3. Effect of indentation load on Vickers hardness.

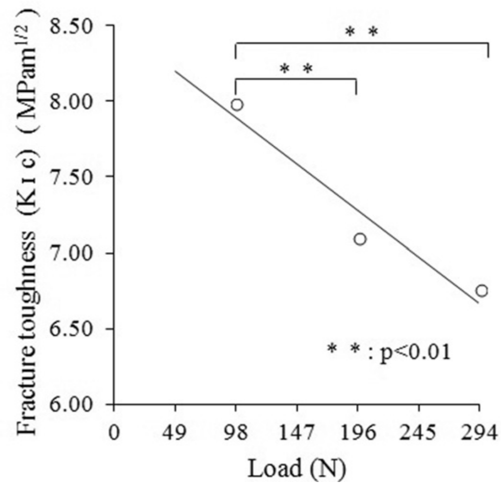

Fig. 4. Effect of indentation load on fracture toughness.

result, 196 and 294 N that had 384 and 520 μm crack lengths, respectively, fell within the recommended range.

Then, only 9.8 N was a high value at calculated Vickers hardness as shown in Fig. 3, and there were no significant difference at 49, 98, 196 and 294 N. Thus, Vickers hardness is calculated high value when the 9.8 N of indentation fracture load is too light because indentation becomes small to not enough impressed. At fracture toughness values, 9.8 and 49 N cannot be calculated because the cracks were not occurred. There was no significant difference between 196 and 294 N in calculated 98, 196 and 294 N.

Therefore, at 9.8 and 49 N are too light in measuring fracture toughness value of zirconia by IF method. We concluded that 196 and 294 N were optimum conditions because measuring results are stability, also size of indentation and crack are within the recommended range.

4.2. Observation of indentations and cracks

When a Vickers indenter is pressed onto the surface of a brittle solid, first, an inelastically deformed region is produced immediately beneath the indenter. When loading is increased and exceeds a certain threshold value, a semicircular vertical crack is generated from the end of the deformed region. This crack grows and reaches to the surface with further increase of loading, and then forms a semicircular or semielliptical crack. No crack from indentation was observed at 9.8 and 49 N of indentation fracture load in this study. However, as shown in Fig. 5, cracks can be observed even at 49 N in SEM observation.

A diamond square pyramid with angles between opposite faces of 136° was clearly observed in all indentations produced by the Vickers indenter in this study; in other words, the shape of the indentations was normal in type and a similar shape with the metal material. In addition, cracks produced from the corners of the square pyramid were fine and linear. A study by Okada et al. [24] sing dental porcelain indicated that the rim around indentation became unclear because of fracture due to chipping at indentation fracture load of 196 N and above, and also indicated a curved shape that is assumed to be produced by chipping and transgranular fracture in observation of cracks. The reason why chipping and fracture observed in dental porcelain was not observed in zirconia at all can be attributable to toughness of zirconia. It is considered that toughening of zirconia is due to stress-induced phase transition, curvature of crack, micro-cracking, and surface compression stress; and that especially transformation mechanism due to stress-induced phase transition has a major contribution [30]. Zirconia produces about 4% of volume expansion by crystal phase transformation that is caused by transformation of tetragonal phase stabilized at room temperature to monoclinic phase by loading. Growth of cracks in the Vickers test starts from the beginning of fracture at the end of indenter, i.e., formation of frontal process zone, and progresses to process zone wake that is expanded frontal process zone, and further to rupture due to the reaching of cracks to the outer surface of the end of the test specimen. Therefore, it is considered that expansion of cracks is prevented because phase transformation from tetragonal phase to monoclinic phase accompanied by volume expansion as well as compression stress at the leading end of the cracks is occurred when cracks are grown. In SEM image of 196 and 294 N in Fig. 5, surface changes are observed around cracks. These correspond to a schematic diagram of suppression of crack development described by Piconi and Maccauro [31] and Ban [32] (Fig. 6). Accordingly, it is considered that phase transition was actually occurred in process zone wake around the developing crack generated by high loading of 196 or 294 N.

4.3. Clinical evaluation

Demands for esthetics and biocompatibility of restoration devices using biomaterials are increasing in patients who want dental therapy. Especially, materials for restoration for crown and bridge are shifting from porcelain-fused-to-metal systems

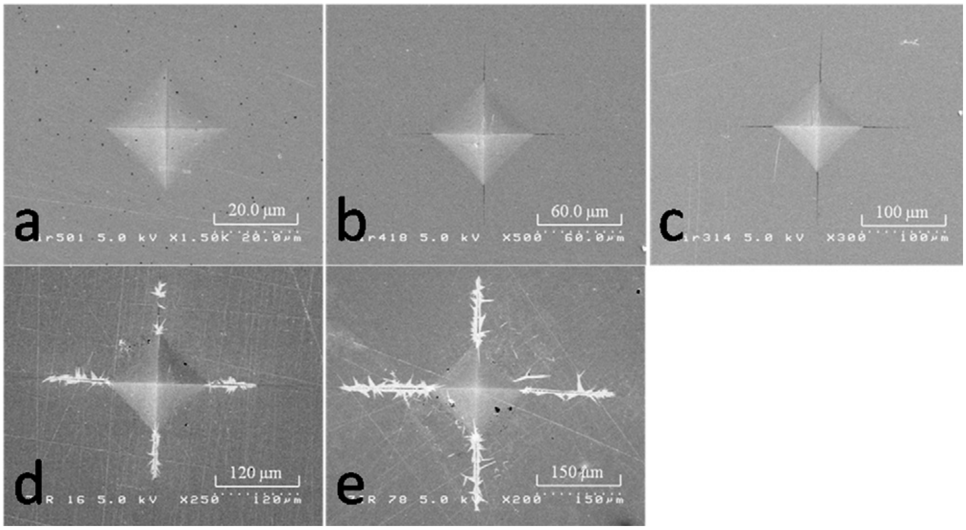

Fig. 5. SEM images of indentation diagonal and crack. (a) 9.8 N, (b) 49 N, (c) 98 N, (d) 196 N, and (e) 294 N.

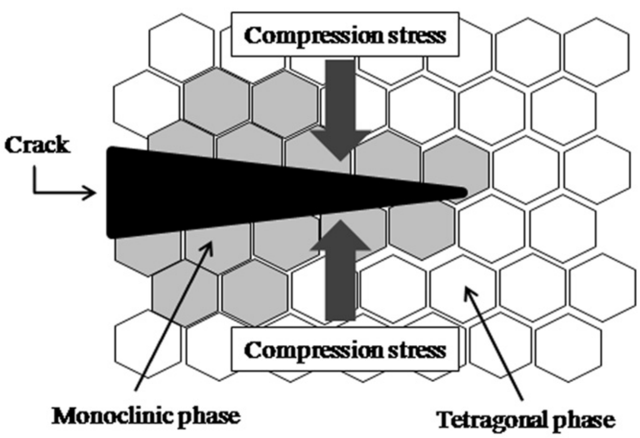

Fig. 6. Schematic diagram of suppression of crack development.

to all ceramic systems. Previously, ceramic restoration has been produced using the refractory cast method, glass-casting method or compression molding method, and is mainly used for inlay, onlay, laminated veneer, and crown. Presently, however, a zirconia frame produced by a CAD/CAM system is applied to a wide range of clinical settings including bridge for multiple tooth defects. A Zirconia frame is used as a high-strength ceramic material with functions as well as, or better than, metal frames that have been used for bridge, and is also clinically applied as abutment for implants and superior restoration devices. Conventional restoration devices made of common dental porcelain have resulted in failure caused by a fracture due to its low fracture toughness value. Therefore, not only bending strength, which is a mechanical strength, but also fracture toughness value should be considered when a new product would be applied to a clinical setting. The fracture toughness value is an especially important measure for zirconia used for bridge frames. Zirconia in this study has high mechanical strength of 6.8–8.0 $MPam^{1/2}$, which is more than double that of existing ceramics, as compared with Pencraft, Vitadur, Cryscera, IPS Empress 2, and In-Ceram Alumina, which have fracture toughness values of 3.11, 1.9, 2.78, 3.02, and 4.7, respectively [33]. In addition, while it is a brittle material, it is indicated that zirconia is a material superior for breakage and fracture that are of most concern in dental brittle materials under the masticatory environment in this study because zirconia showed clear indentation like metal materials, and generated cracks showed a linear shape without chipping and fracture due to stress-induced phase transition.

Because of the emergence of such a material, reliability of metal-free restoration applying zirconia is increasing even in cases that used to require metals such as bridge.

5. Conclusions

Within the limitation of this study, following conclusion is made:

1. Indentation and cracks were observed when the indentation fracture loads were 98, 196 and 294 N, but there was no significant difference between 196 and 294 N. Therefore, optimum experimental conditions of the indentation fracture load in the IF method were determined as 196 and 294 N, and obtained values were 7.1 and 6.8 $MPam^{1/2}$.
2. In SEM observation of the indentation and the cracks, no chipping and fracture were observed, and resulting cracks were clear and extended linearly. In addition, surface changes were observed around the cracks in 196 and 294 N.

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